Formation of Polar Stratospheric Clouds on Preactivated Background Aerosols

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Polar stratospheric clouds (PSCs) play an essential role in ozone destruction in the Antarctic and Arctic. Heterogeneous react ions occurring on PSC surfaces promote the release of active chlorine and deactivate nitrogen oxides; sedimentation of PSC particles leads to ix-reversible removal of HNO_3 from the stratosphere (denitrification), which critically determines the photochemical ozone depletion efficiency of Cl radicals. Despite their importance, the formation mechanism of PSCs remains uncertain. We present the results of laboratory simulations of the growth of nitric acid trihydrate, $HNO_3 \cdot 3H_2O$, on sulfuric acid tetrahydrate, $H_2SO_4 \cdot 4H_2O$. The observations reveal that under typical stratospheric conditions uptake of HNO_3 on a $H_2SO_4 \cdot 4H_2O$ substrate results in a surface coverage of approximately one monolayer or less, and that initial $HNO_3 \cdot 3H_2O$ nucleation requires a large supersaturation. We also observe that a $H_2SO_4 \cdot 4H_2O$ substrate, onto which a $HNO_3 \cdot 3H_2O$ film has been deposited and subsequently evaporated, exhibits a remarkable enhancement in its nucleation ability for this nitric acid hydrate. In the stratosphere, PSC particles may experience repeated cycles of evaporation and condensation of HNO_3 on preexisting background frozen sulfate aerosols. Hence, growth of $HNO_3 \cdot 3H_2O$ on preactivated aerosols provides one important mechanism for polar stratospheric cloud formation.

1. 1995 Fall Meeting

2.30002034 (AGU member)

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5. (a) A03 Stratospheric Chemistry and Dynamics

(b) 0305-Aerosols and Particles
0340-Middle Atmosphere-Composition and Chemistry
0370-Volcanic Effects

(c) Climate and Global Change

6. No

7. No

8. Invoice $60 to Attached P.O. # xx at Jet Propulsion Laboratory, 4800 Oak Gorve Dr., Pasadena, CA 91109

9. C

10. No

11. No